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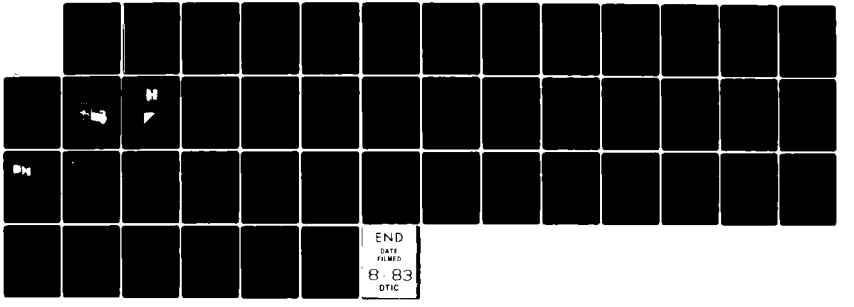

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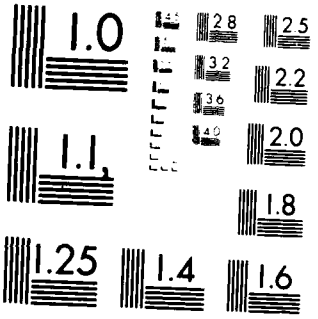

INVESTIGATION OF OPTICAL FIBERS FOR NONLINEAR OPTICS

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In the first year of this program a significant portion of these objectives has been achieved. Differential thermal analysis (DTA) has been performed on KDP, AgCl, $LiIO_3$ and (on a concurrent contract) KRS-5. Growth rates for KTN, $KNbO_3$ and $LiIO_3$ were established in carefully controlled experiments on bulk crystals. AgCl was identified by DTA and chemical purification studies using zone refining in a CCl_4:He atmosphere as a good test material for SC fiber fabrication by a "traveling zone" method. This approach produced fibers more than 2 cm in length and appears to be very promising for certain congruent materials. KDP fiber growth was attempted in a Bridgman apparatus and showed limited success, producing encapsulated fibers roughly 2 cm long. The potential of Czochralski growth methods for KTN and $KNbO_3$ fibers was also evaluated.

In addition to crystal growth and fiber fabrication efforts, an alternate approach to nonlinear optical fibers was developed and shown to have significant potential. Experiments demonstrated that high quality glass fibers could be smoothly incorporated into nonlinear media to permit coupling the evanescent field of guided light to the crystal of interest. Light in the guide can generate phase-matched, nonlinear polarization in the surrounding medium which radiates new frequencies back into the guided mode. Two methods successfully produced oriented, embedded fibers in $LiIO_3$. Numerous device applications are envisioned for hybrid fibers of this type. This unique approach has provided a new avenue for nonlinear fiber fabrication at a time when it has become clear that no one method will be satisfactory for all nonlinear material of interest. Significant differences in physical and chemical properties continue to dictate the adaptive approach to methodology originally proposed for this program.

PREFACE

This is the first annual report describing our progress on the development of optical fibers for use in nonlinear optical systems. The one-year period covered is from 1 January 1982 to 31 December 1982.

Participating in the research program are the following personnel: Larry G. DeShazer as principal investigator; J.A. Harrington, A.C. Pastor, and R.C. Pastor in fiber fabrication, and S.C. Rand in hybrid single crystal fiber studies.

This research is being supported by the U.S. Air Force Office of Scientific Research, Bolling AFB, D.C. The program is under the technical supervision of Dr. Howard Schlossberg, Physics Directorate.

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SECTION 1

RESEARCH OBJECTIVES

The main objective of this contract is to find methods of fabricating single crystal nonlinear optical fibers. Successful techniques are to be utilized to make crystal fibers that will be useful for nonlinear devices, particularly in the infrared. Device concepts and applications are also to be formulated. This work has been divided into three parts which are described below.

Task 1 entails the measurement of physical and chemical properties of linear and nonlinear materials relevant to the preparation of single crystal and nonlinear fibers. Differential thermal analysis (DTA), vapor pressure analysis and thermogravimetric methods (TGA) are employed for this purpose. The emphasis is on selection of fiber growth methods most appropriate for particular materials or classes of materials. The scope of methods investigated is intended to be broad in order to encompass at least three different candidate materials.

Under task 2 a variety of methods for production of single crystal fibers are to be developed, recognizing that no one method can be applied even in principle to SC fiber growth of all materials of interest. Efforts will be directed toward, but not limited to, the study of:

- Traveling Zone Method - A method for conversion of polycrystalline fibers to SC fibers. This technique uses a small heater to recrystallize small sections of extruded polycrystalline fiber which pass through a hot zone on rollers. Long lengths are possible in principle for congruent materials.

- Vapor-Stabilized Bridgman Method - A Bridgman method of producing single crystal fibers in which the melt is vapor-pressure stabilized. This method will be applicable to nearly congruent melters like KDP. When operating parameters have been established its

applicability to isomorphous compounds such as ADP and deuterated forms such as KD*P will be assessed.

- Inverted Czochralski Method - A method for producing single crystal fibers. This method makes use of a growth orifice below the melt along with the stabilization of melt composition. Dynamic injection of makeup material or provision of a reservoir in melt contact with the main charge (but held at a slightly higher temperature) is the basis for stabilization.

- Capillary-Fed Czochralski Method - A method for congruently melting materials. Capillary designs can overcome thermal steady state limitations of the conventional Czochralski technique applied to fiber growth. By reducing the ratio of free surface of the melt to growth interface, thermal problems can be minimized and surface tension may be useful in providing mechanical stability at the growth interface.

- Hybrid Single Crystal Fiber Method – Develop a hybrid single crystal fiber. In the presence of good optical contact between a glass fiber core and a bulk, nonlinear crystal, the evanescent portion of the guided wave can couple to nonlinear polarization in the crystal. This results in a nonlinear hybrid fiber with properties controlled by fiber diameter and orientation relative to crystal axes.

Task 3 is concerned with the measurement of optical properties of the fabricated fibers and development of device applications. Characterization of absorption and scattering losses as well as observation of nonlinear optical effects constitute the core of this part of the program. Measured properties are to be analyzed using existing theories of nonlinear processes and light scattering in fibers (particularly from surface imperfections). New theoretical approaches are to be developed only if necessary for analysis. Primarily in the second year of the program this task will include consideration of new devices using nonlinear fibers.

SECTION 2

STATUS OF THE RESEARCH EFFORT

A. INTRODUCTION

During the first year of the Optical Fibers for Nonlinear Optics program several promising methods for single crystal fiber growth have emerged, and important limitations of these and other methods have been identified. Single crystal (SC) fiber fabrication of AgCl and KDP achieved by the Traveling Zone and Bridgman methods is very encouraging. Results with these approaches are described in subsections B and C, respectively, of this section. In subsection D a low growth rate of the Inverted Czochralski method for incongruently melting crystals is discussed, and in subsection E a Capillary-fed Czochralski approach is shown to hold considerable promise for congruent crystals. In subsection F experimental results with $LiIO_3$ are used to demonstrate that Hybrid SC Fibers provide a novel, versatile method of fabricating oriented single crystal fibers based on virtually any ionic nonlinear crystal.

It should be emphasized that as a class nonlinear optical materials encompass the entire range of thermal behavior possible in compounds. It has therefore become increasingly clear in this program that no one method of SC fiber growth can accommodate the disparities of all the crystals listed in Table I. Hence it must be recognized that fabrication of SC fibers from these materials necessarily calls for a variety of methods. Our progress can be directly linked to this perspective on the problem and the broad scope of our efforts.

Table I. Classification of Nonlinear Crystals Based on Thermal Behavior

Thermal Classification		Nonlinear Optical Crystals
Congruent Melting		$LiNbO_3$, $LiTaO_3$, BSO, $Gd_2(MoO_4)_3$, $Tb_2(MoO_4)_3$, BeO, CuBr, CuCl, CuI
Incogruent Melting		
(a)	Solid Solutions	KTN, $Ba_2NaNb_5O_{15}$, $PbNb_4O_{11}$, $K_3Li_2Nb_5O_{15}$, $K_{0.8}Na_{0.2}Ba_2Nb_5O_{15}$, $Sr_{0.5}Ba_{0.5}Nb_2O_6$
(b)	Peritectic Compounds	$KTaO_3$, $KNbO_3$, $PbTiO_3$
Thermally Unstable Materials		
(a)	Chemical Change below melting	$LiIo_3$, KDP, $AgGaS_2$, $ZnGeP_2$, CdSe, CdS, SiC, $K_2C_4H_4O_6 \cdot 1/2\ H_2O$, $(NH_2CH_2COOH)_3 \cdot H_2SO_4$ $LiCHO_2 \cdot H_2O$, $(NH_4)_2C_2O_4 \cdot H_2O$, α-HIO_3, Ag_3AsS_3, Ag_3SbS_3, ADP, AD*P, CsH_2AsO_4, KH_2AsO_4, $BeSO_4 \cdot 4H_2O$, KD*P, RDA, RDP, ZnO, ZnS, CdTe, GaSb, GaAs, GaP, InSb, InAs, InP, ZnSe, β-ZnS, ZnTe, $NaBrO_3$, $NaClO_3$
(b)	Polymorphic below melting	$NaNO_2$, α-SiO_2, α-HgS, $BaTiO_3$.

B. TRAVELING ZONE METHOD FOR SC FIBER GROWTH

The traveling zone method of fabricating SC fibers is unlike any other fiber growth technique. In this method we begin with a polycrystalline (PC) fiber made by a hot extrusion method (Ref 1). The fiber is then placed between two pairs of drive rollers, as shown in Figure 1. A small heater coil is used to melt a tiny section of the fiber, and as the fiber travels through the heat zone, the PC fiber is converted to SC fiber.

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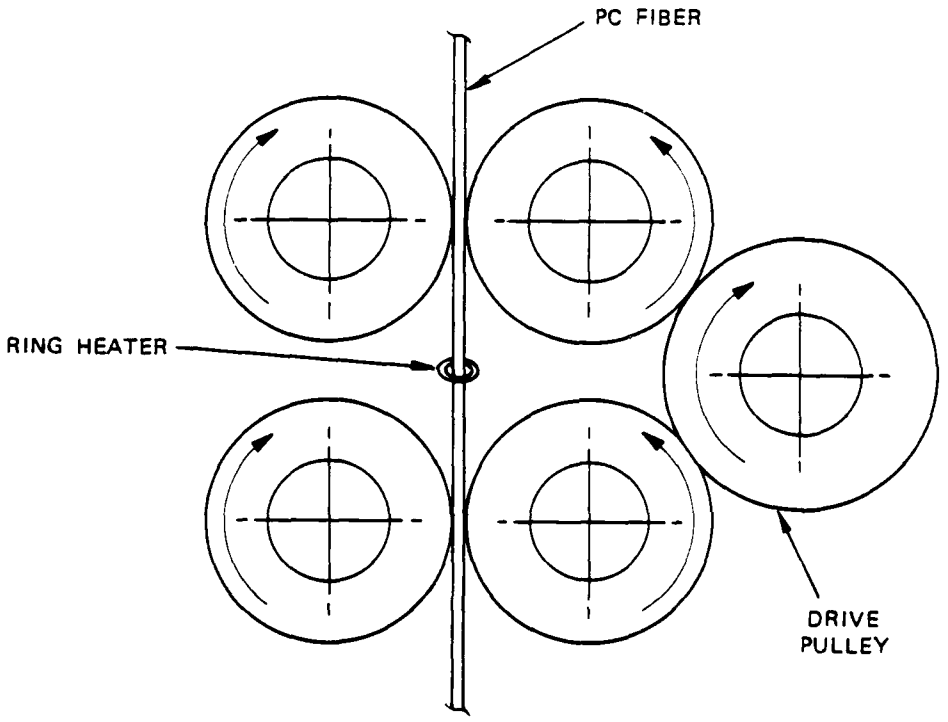


Figure 1. PC to SC fiber conversion for unencapsulated fibers. Only a small portion of the fiber is melted in the ring heater.

The traveling zone technique has the unique advantage of beginning with PC fiber and converting it to SC fiber. In addition, the fiber surface is not in contact with any shaping element, and thus the surface quality should be very good. The method also permits some reduction in the final SC fiber diameter, although a 1:1 fiber diameter ratio appears to be optimal.

The fiber pulling apparatus for growing SC fiber is shown in Figure 2. As shown in the insert, the PC fiber, moving either up or down in the critical melt zone, is converted to SC fiber. The fiber is guided by two quartz capillary tubes located at the top and bottom of the rollers. To minimize convection currents, we have also made a small quartz enclosure (not shown in Figure 2) which encompasses the heater and melt zone. This small enclosure also has capillary guide tubes at the top and bottom to keep the fiber precisely aligned within the heater coil. Using the apparatus shown in Figure 2, we are able to control accurately the speed of the fiber, the temperature of the heater coil, and provide a melt zone from one to three fiber diameters in length. Generally, we operate with a melt zone approximately one fiber diameter in length.

The initial choice of fiber material was AgCl. This material was chosen because it is easy to extrude into PC fiber. Its vapor pressure is low at the melting point and the melt (yellow in color) is readily distinguished from the solid state (which is clear) allowing the profile of the melt zone to be studied carefully. After we perfect our techniques with AgCl fiber we will attempt more difficult crystals such as $LiNbO_3$.

The results for SC fibers of AgCl are shown in Figure 3. In Figure 3(a) the fiber is shown in place between the guide tubes, together with the heater coil, during operation. An approximately 2-cm-long section of SC fiber is shown on top of a scale in Figure 3(b). While only short segments of SC fiber have been made, we do not see any obstacles to making SC fibers 1 m or more in length using the existing apparatus. At a later time it

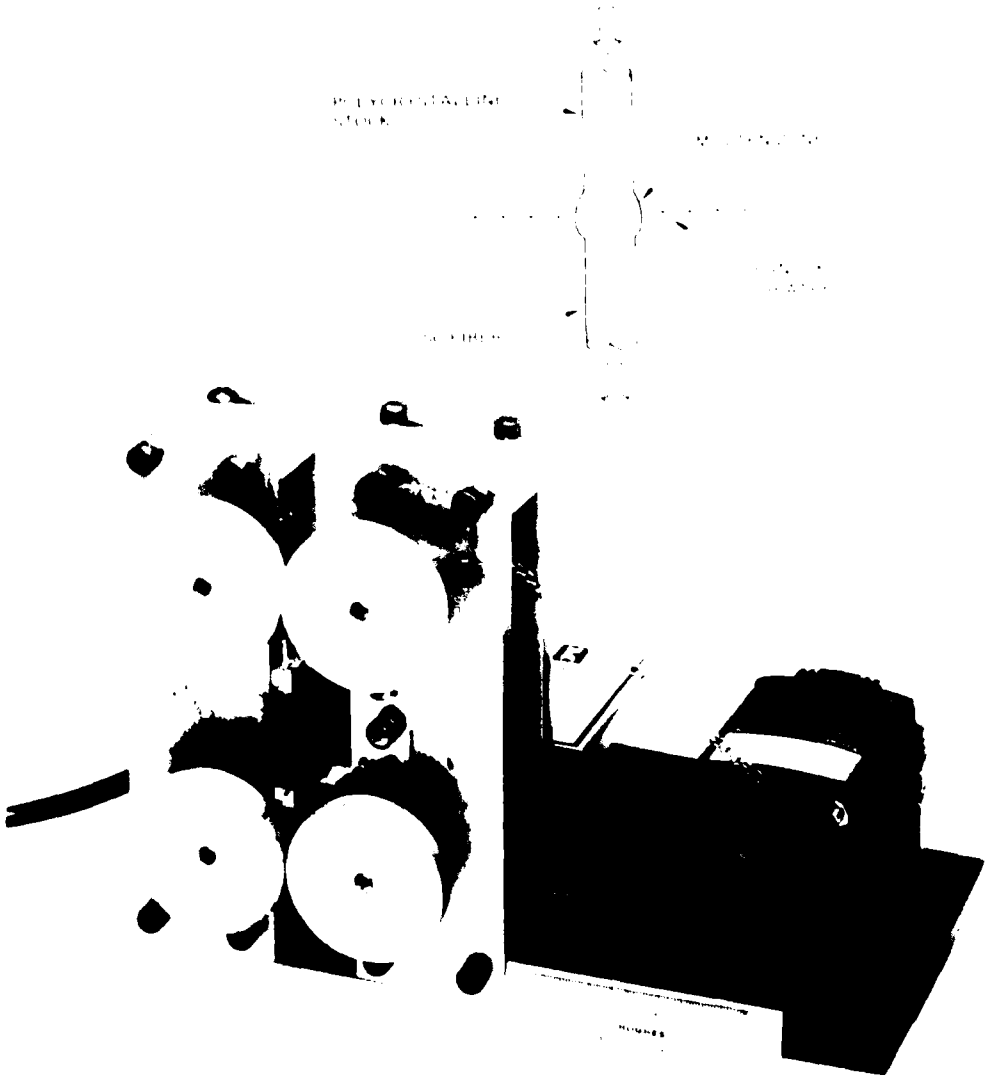
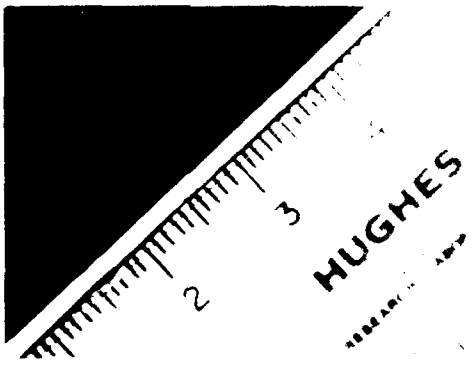


Figure 2. Fiber-pulling apparatus for feeding polycrystalline AgCl fiber through heat zone for conversion to single crystal fiber. The insert shows details of the critical molten zone.

(a)



(b)

Figure 3. SC fiber of AsCl prepared by the travelling-zone method. (a) 500-μm diameter fiber between two guide tubes showing molten zone and heater coil. (b) A section of SC fiber resting on scale divisions.

will be necessary to add a feedback system to control fiber diameter, an inert or reactive atmosphere to prevent impurity contamination of the fibers, and a method to seed the fibers for growing oriented SC fibers.

C. VAPOR-STABILIZED BRIDGMAN METHOD FOR SC FIBERS

On the basis of differential thermal analysis (DTA) studies at HRL, KDP (potassium dihydrogen phosphate) has been selected to be the test material for this method of SC fiber growth. A DTA thermogram of KDP is shown in Figure 4. It reveals changes in the constitutional water upon heating and exhibits a rather poorly defined melting transition above 260°C. Remarkably, although the material evidently melts incongruently, the DTA shows a sharp exothermal peak suggestive of crystallization of a congruent melter upon cooling. It is this feature which suggests that vapor-stabilized Bridgman growth can be a successful technique for KDP fibers.

Figure 5 shows the loading and sealing of the HRL-designed crucible preparatory to SC growth in the Bridgman mode. The last diagram in the sequence gives a faithful representation of the apparatus constructed for the KDP experiments. Our initial Bridgman runs with KDP charges yielded short segments of clear fiber roughly one inch long. X-ray analysis indicated that the crystal structure in these clear regions corresponded to either $K_4H_6P_4O_{15}$ or $3KH_2PO_4 \cdot KPO_3$. The DTA and x-ray evidence jointly suggest the following chemical reactions:

$$4KH_2PO_4(s) \xrightarrow[\text{heating}]{\sim 270\,^\circ C} 3KH_2PO_4 \cdot KPO_3(\ell) + H_2O(g)$$

$$\Big\downarrow \text{ cooling to room temperature}$$

$$3KH_2PO_4 \cdot KPO_3(s) + H_2O(\ell) \; . \qquad (1)$$

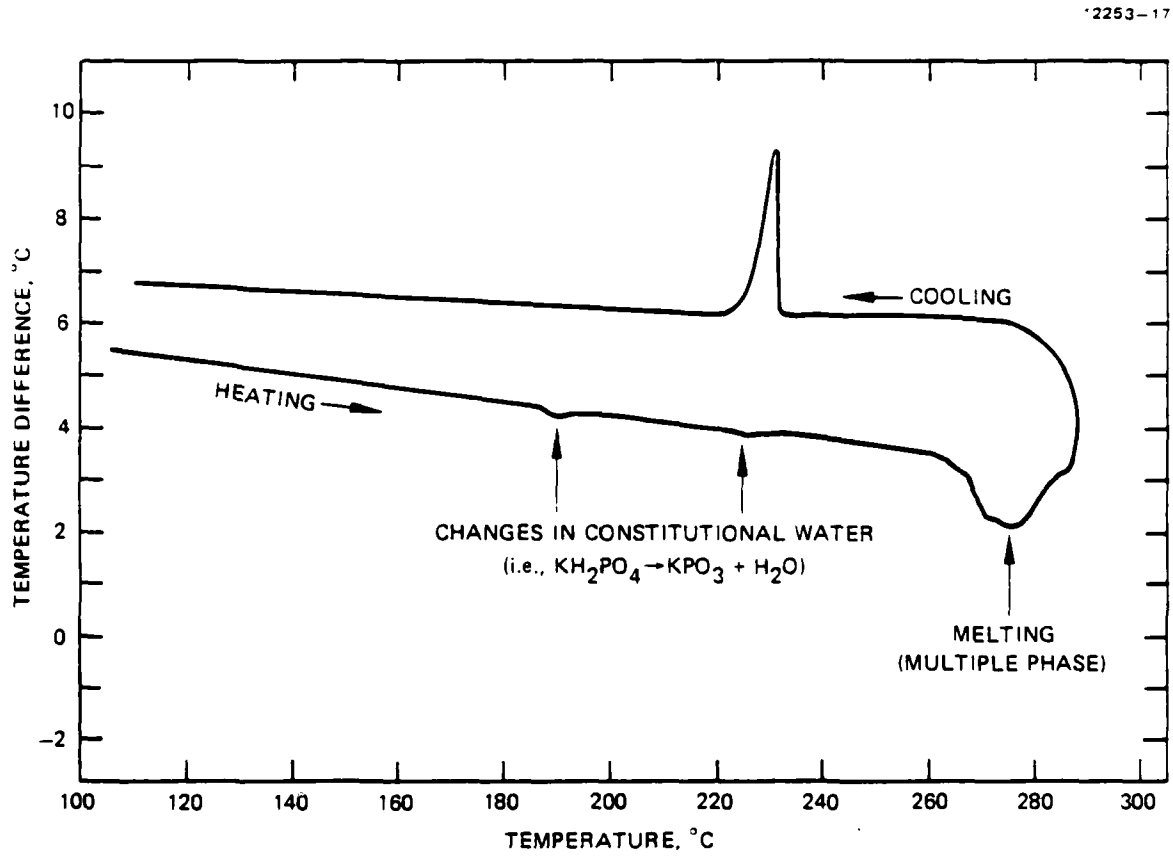


Figure 4. Differential thermal analysis curve for encapsulated KDP.

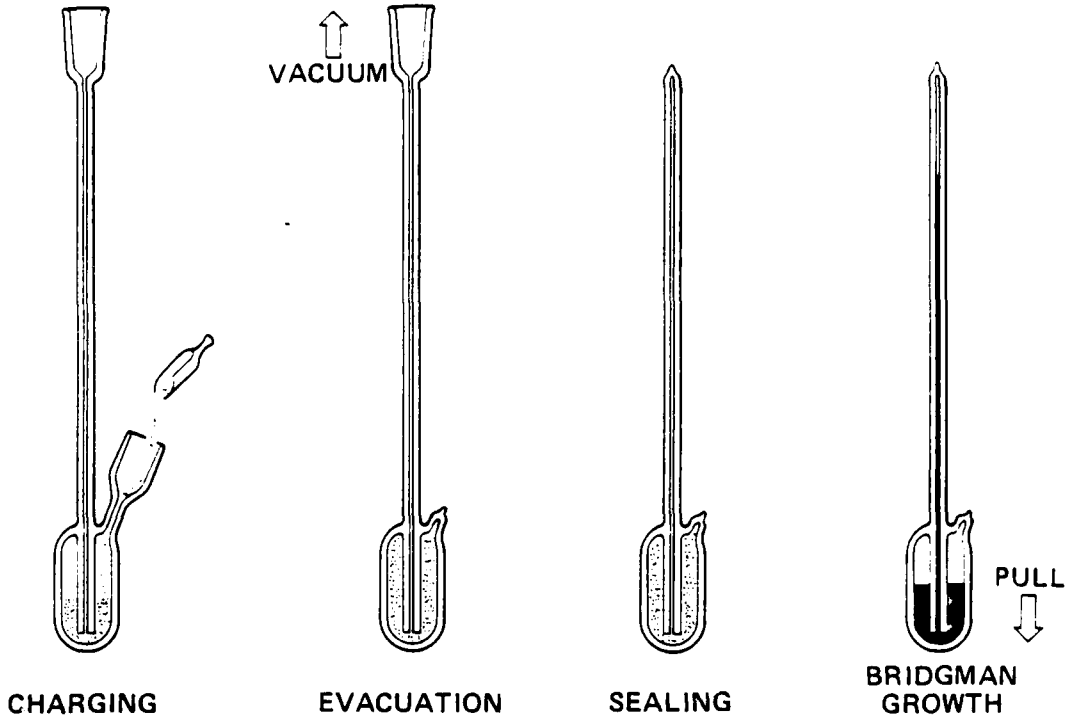


Figure 5. Stabilized Bridgman growth apparatus for SC fibers of KDP. The figure illustrates the sequence of loading, evacuation, sealing and crystal growth.

Growth of KDP can evidently be effected if the first step of the chemical reaction is made reversible. The DTA and x-ray results have led us to speculate that the melting of KDP will be rendered reversible if the process is carried out under a high water vapor pressure. For growth from the melt to be feasible, the reaction must be

$$KH_2PO_4(s) \underset{}{\overset{\text{high } p(H_2O)}{\rightleftharpoons}} KH_2PO_4(\ell) \ . \qquad (2)$$

The appropriate experimental approach is to determine the threshold value of the partial pressure of water, namely, $p(H_2O)$, which will make KDP melt reversibly.

For this purpose a stainless steel bomb with fused silica liner has been designed and fabricated at HRL for Bridgman operation with H_2O overpressure (Figure 6). Initial runs will be aimed at bulk SC growth. Success with bulk growth in this configuration would in itself be a significant achievement since all commercial fabrication of KDP is performed exclusively from solution at the present time. Extension of the method to fiber growth will be accomplished by the introduction of fused silica capillaries into the melt region of Figure 6.

D. MELT-STABILIZED INVERTED CZOCHRALSKI METHOD FOR SC FIBERS

An analytic study has revealed limitations of the process of SC growth of an incongruent melter from a fluxed melt. There are two cases of melting incongruency to consider; each is treated in turn in this section.

The first is exemplified by peritectic compounds like $KNbO_3$ which crystallize below the peritectic temperature from a finite melt. As such compounds are cooled, the phase that crystallizes has an unvarying composition although the melt composition changes, becoming progressively leaner in the peritectic compound as the temperature decreases. Potassium niobate ($KNbO_3$) has a peritectic temperature of 1039°C but can be melted below this temperature by the addition of excess K_2O.

Our analysis of peritectic growth from the fluxed melt yields the equation for the linear growth rate:

$$R = D(dX(\ell)/dT)(dT/dZ)(V_s/V_\ell X(s)) \ , \qquad (3)$$

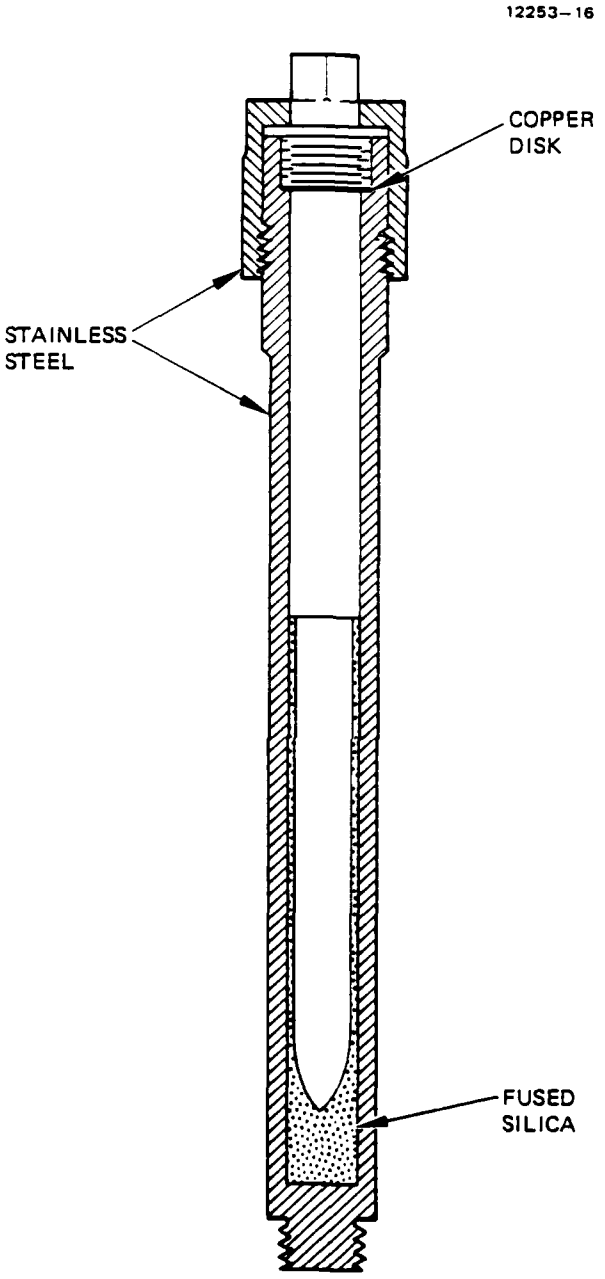


Figure 6. Crystal growth bomb for KDP preparation in the presence of H_2O overpressure.

where D is the mass diffusion coefficient of the crystallizing species in the fluxed melt, $dX(\ell)/dT$ is the reciprocal of the slope of the liquidus in the binary phase diagram of crystallizing species versus flux, dT/dZ is the temperature gradient in the melt at the crystal growth interface, the V's are molar volumes, and $X(1)$ is the liquid mole fraction of the crystallizing species. Order of magnitude values of these factors for $KNbO_3$ are $D \sim 10^{-9}$ m^2/s, $dX(\ell)/dT \sim 10^{-3}$ °C^{-1}, $dT/dZ \sim 10^{3}$ °C m, $V_s/V_\ell \sim 1$, and $X(s) \sim 1$. Substitution of these values into (3) gives $R = 10^{-9}$ m/s, or 0.1 mm/day. This low value has been corroborated by experimental studies undertaken during the year to measure bulk SC growth of $KNbO_3$. Our measurements yielded a value of R = 0.3 to 0.5 mm/day for $KNbO_3$.

The second case of melting incongruency is that of the solid solution in which crystallization occurs from a melt of differing composition. As the mixed system crystallizes, the composition of the finite melt changes. Therefore, the temperature has to be lowered for crystallization to continue; but the next crystallizing layer will have a composition differing from that of the preceding crystallized layer since it is crystallized from a melt of changed composition. For the crystal to be homogeneous, time must be allowed for the preceding crystallized layer to change its composition to match that of the next crystallizing layer. Since the mechanism for such a change is diffusion in the solid, it is very slow. Homogeneous crystal growth of a solid solution from a finite melt is bottlenecked by solid state diffusion.

Our analysis of this process yielded the equation for the linear growth rate,

$$R = D(dX(s)/dT)(dT/dZ)/X(s) \qquad (4)$$

where D is now the mass diffusion coefficient for the solid, $dX(s)/dT$ is the reciprocal of the slope of the solidus, and dT/dZ is the temperature gradient in the solid at the interface.

In the solid, D is typically 10^{-6} to 10^{-5} times D in the liquid, and there is no order-of-magnitude change in the other factors. Therefore, R = 10^{-6} mm/day is an optimistic value for this case.

The melt-stabilized inverted Czochralski growth method proposed by us in effect makes the size of the melt infinite, so that for $KNbO_3$ R will be similar to the calculated value for peritectic growth. The driving force of flow of melt through the capillary orifice in this growth method is the difference in pressure between the vapor space inside the vessel containing the melt and that on the outer side of the orifice plus the hydrostatic head due to the column of melt over the orifice (assuming that the growing fiber is externally supported). Hence the maximum growth rate expected for this approach is given by (3). For $KNbO_3$ the fiber growth rate is therefore limited to roughly 0.5 mm/day, making it impractically slow. However, applications of the technique to congruent materials still need to be investigated.

E. CAPILLARY-FED CZOCHRALSKI METHOD FOR SC FIBERS

The capillary-fed Czochralski method of SC fiber growth was invented at HRL to overcome some of the thermal and mechanical problems inherent in conventional Czochralski methods. The capillary modification is intended to handle congruently melting materials, which behave as one-component systems under crystallization conditions.

A discussion of the concomitant changes in the material processing is helpful at this point, particularly as they relate to fiber production. The solid and molten states of congruent materials coexist at the melting point and the growth interface is obviously at that melting point at every instant during the growth of a crystal from the melt. What is not so evident is that a finite layer of the melt adjacent to the interface must be at the melting point also, with no sensible heat flowing

through it in either direction. Otherwise, growth will be interrupted. During crystal growth, the growth interface is to be regarded as a heat source because each growth layer that deposits on it gives off its latent heat of crystallization in the process. This heat has to be withdrawn from the interface and conducted away from the melt at a rate matching the growth rate of the crystal. Therefore, a state of mutual thermal stabilization exists between the melt and the crystal at the growth interface. In Czochralski growth the melt also interfaces with the gaseous surroundings. A thermal steady state has to be established at the common periphery of the phases before steady state growth is feasible. This thermal steady state is difficult to achieve if the ratio of free surface of the melt to growth interface is made large. On the other hand, as this ratio is reduced small surface tension effects due to the free surface of the melt are enhanced and, particularly in the case of SC fiber growth, mechanical equilibrium becomes an added consideration.

The capillary-fed Czochralski method helps overcome the difficulties in SC fiber growth inherent in the conventional Czochralski method. Apparatus constructed for initial experiments at HRL is shown in Figure 7. A capillary tube suspended in a crucible in effect reduces the crucible diameter to the desired diameter of the fiber and brings surface tension into play. It is assumed that the melt wets both the crucible and the capillary tube walls. Capillarity has been found to maintain the level of melt in the capillary, since its height is less than that of the capillary column that can be supported by the surface tension of the melt. The melt does not overflow because the tube does not have capillary dimensions at the growth nozzle shown in Figure 7. We believe the nozzle angle is critical in steady-state fiber growth. Current experimental work is directed toward optimization of this nozzle parameter, taking surface tension fully into account.

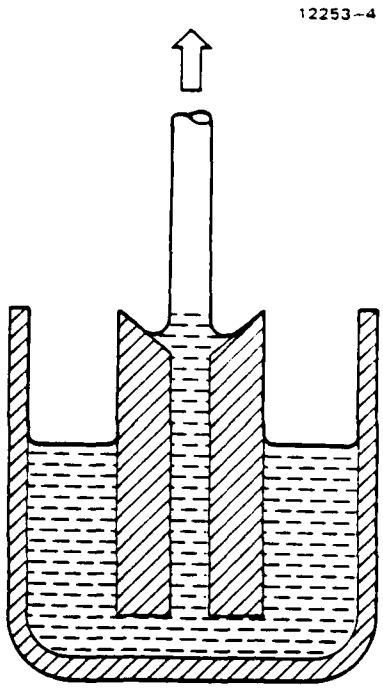


Figure 7. Capillary-fed Czochralski apparatus for growth of SC fibers.

F. HYBRID SINGLE CRYSTAL FIBERS

One of the key objectives of the present nonlinear fiber program is to survey growth methods and identify the most successful techniques for preparation of particular nonlinear materials or classes of materials. However, it is anticipated that even with the best techniques it will be difficult at first

to achieve the surface quality and uniformity routinely available with glass fibers. Because the efficiency of phase-matched processes in nonlinear fibers is sharply reduced by surface imperfections and dimensional irregularities of the guide, we have developed a unique, alternative approach to nonlinear fiber fabrication which avoids this problem and lends itself to early development of device applications.

The new approach combines the desirable uniformity of glass fibers with bulk nonlinear crystals in a hybrid configuration. As depicted in Figure 8, hybrids operate as nonlinear optical fibers by virtue of the coupling between the evanescent portion of guided waves and the nonlinear polarization of bulk, noncentrosymmetric crystals. Unclad fiber cores can be incorporated

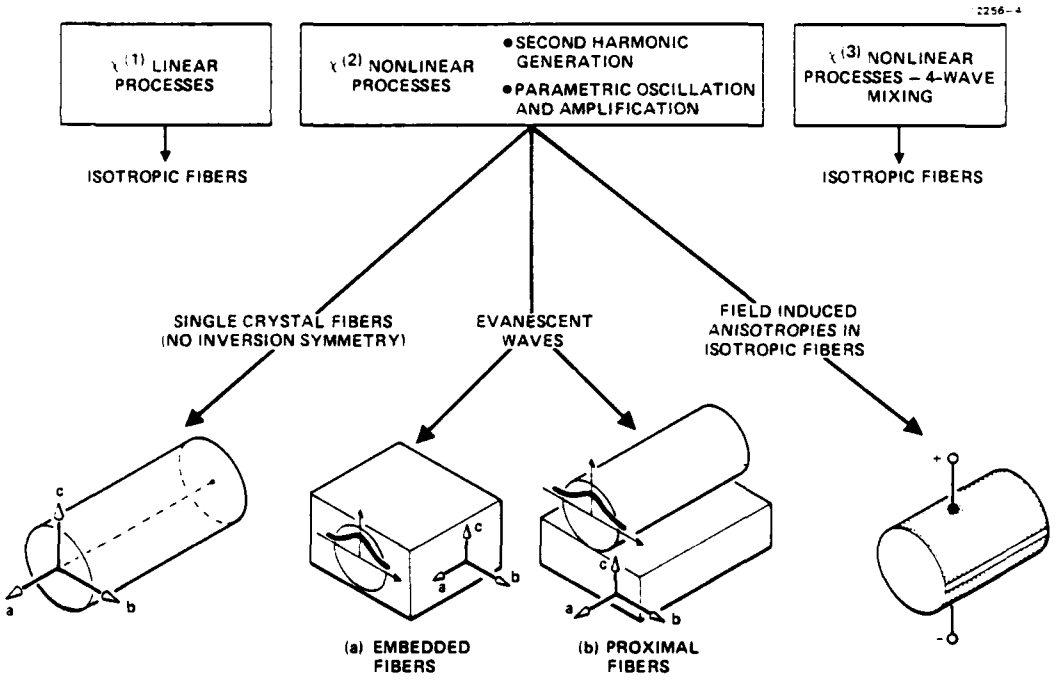


Figure 8. Methods of fabricating second order nonlinear fibers.

in high quality crystals by two techniques which yield excellent optical contact between fiber and crystal: the "saturated solution press" and "growth embedding" techniques. In the former method a fiber can be smoothly pressed into a crystal surface to a depth roughly equal to its diameter by displacing the chemical equilibrium,

$$A\ [\text{Solid}] \underset{k_r}{\overset{k_f}{\rightleftharpoons}} A\ [\text{Solution}]\ , \qquad (5)$$

to the right. According to LeChatelier's Principle (Ref. 2), for all systems in which the molar volume of the solid exceeds that of the solution, simple application of pressure will cause the equilibrium constant, $K = k_f/k_r$, to increase. Hence, local regions of the solid go into solution when subjected to point or line pressure. The application of pressure to a fiber at such a crystal surface in solution therefore causes the fiber to "submerge" into the solid. In the second method a fiber is totally encased by the crystal during growth from solution.

Results of experiments with these embedding techniques are shown in Figures 9 and 10. $LiIO_3$ has a calculated negative volume change of 17.4 percent for the forward reaction; hence, it was selected for the initial work. The photographs demonstrate the good optical contact achievable in a few hours between a glass fiber core 125 μm in diameter and a $LiIO_3$ crystal in a 90° phase-matching geometry.

Because the fibers can be arranged to lie in any direction with respect to crystal axes, our embedding techniques are versatile ways of achieving oriented, nonlinear optical fibers with controlled properties (see Section 5 on Invention Disclosures). For example, by choosing the appropriate fiber orientation and diameter we should be able to make use of any specific electro-optic coefficient for second harmonic generation in any range

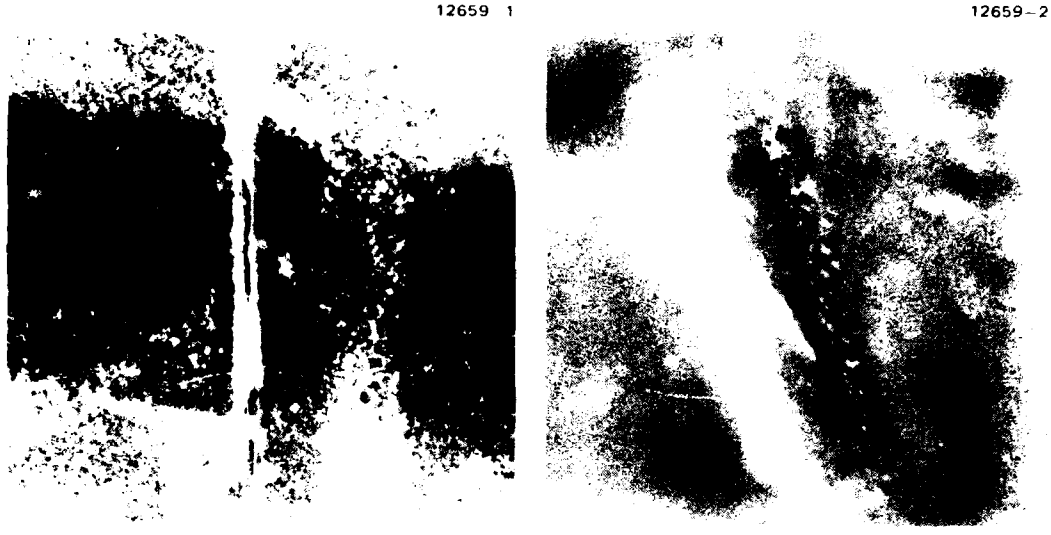


Figure 9. Hybrid single crystal fiber produced by the saturated solution press method. (a) Silica fiber core (125 μm diameter) embedded in surface of $LiIO_3$ in conventional 90° phase matching geometry. (b) 100X magnified view of the embedding trough after removal of short section of fiber.

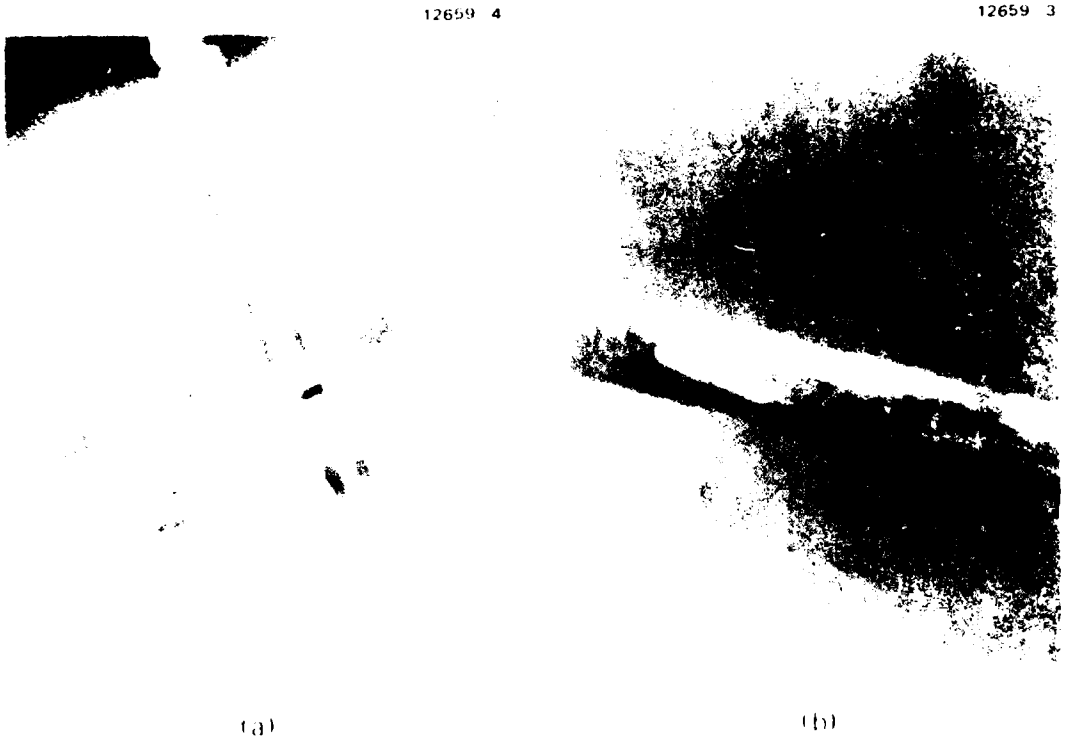


Figure 10. Hybrid single crystal fiber produced by the "growth embedding" technique. (a) Silica fiber core (1.5 μm diameter) extruding from $LiIO_3$ single crystal in conventional 90° phase matching geometry. (b) 100X magnification of encapsulated fiber viewed down optic axis through such a cut, polished crystal surface.

free of crystal absorptions. The only condition which must be satisfied is that the propagation constant, $\beta(\omega)$, at frequency, ω, must satisfy

$$2\beta(\omega) \neq \beta(2\omega) . \qquad (6)$$

We therefore see no obstacles in the application of these hybrid fibers to generate second harmonic radiation for the

first time in fiber guides and to improve on the low efficiencies demonstrated previously for this process in planar waveguides (Ref. 3). By exploiting the length advantage of fibers we may be able to realize practical devices for doubling, summing and differencing of cw dye and semiconductor lasers in waveguides as well as parametric oscillators and amplifiers.

SECTION 3

PUBLICATIONS

1. J. A. Harrington, "Crystalline Fiber Optics," in Encyclopedia of Materials Science and Engineering (Pergamon Press, 1982). A copy of this article is provided in Appendix A.

SECTION 4

BIOGRAPHIES OF KEY PERSONNEL

The biographies of the personnel who are contributing to the program are presented in the following pages. L. G. DeShazer is the Principal Investigator of this research program.

LARRY G. DeSHAZER, Head, Laser Optical Materials Section, Hughes Research Laboratories.

Education

B.S. (Physics), University of Maryland, 1956;
Ph.D. (Physics), The Johns Hopkins University, 1963.

Experience

Dr. DeShazer has 23 years of professional experience in the areas of solid-state lasers, nonlinear optics, laser damage physics and atomic spectroscopy. From 1963 to 1966, he was at Hughes Aircraft Company, Culver City, involved in research on energy transfer in solid-state laser materials and studies of laser mode selection techniques.

In 1966 he joined the Physics and Engineering Faculty at the University of Southern California as Associate Professor. He was mainly involved in spectroscopy of rare-earth ions in laser materials and pioneered in the measurement of self-focusing parameters by the optical pulse shaping technique and laser damage to thin dielectric films.

In 1973 Dr. DeShazer was appointed the first Director of the Center for Laser Studies at USC, an applied research organization serving as an interface between industry and university. While on leave of absence from USC, from 1975 to 1976, he was Liaison Scientist with the U.S. Office of Naval Research, London. He left USC in 1978 to return to Hughes at the Research Laboratories with responsibility in solid-state laser and nonlinear optics research programs.

Honors

Fellow of Optical Society of America, Gilman Fellow at Johns Hopkins University (4 years), Phi Kappa Phi, Phi Eta Sigma, Sigma Pi Sigma, Gamma Alpha.

Professional Societies

American Physical Society, Optical Society of America, The Royal Institution of Great Britain, Sigma Xi, IEEE Group of Electron Devices, SPIE.

Publications

Dr. DeShazer is the author of 82 technical papers in the fields of solid-state lasers, nonlinear optics and spectroscopy. He has 3 patents in solid-state lasers, and was the dissertation chairman for twenty Ph.Ds.

JAMES A. HARRINGTON, Senior Staff Physicist, Optical Physics Department, Hughes Research Laboratories.

Education

B.S. (Physics), Grinnell College, 1964;
M.S. (Physics), Northwestern University, 1966;
Ph.D. (Physics), Northwestern University, 1970.

Experience

Dr. Harrington has 13 years of research experience in the area of optical properties of solids. Since joining Hughes Research Laboratories in 1977 he has been involved in the study of low-loss materials and coatings for use as high-power infrared laser components. Recently, he has been appointed manager, infrared fiber optics responsible for coordinating all IR fiber programs. His studies of IR fiber materials include measurement of ultimate absorptive and scattering losses in transparent materials. Prior to joining Hughes, he was assistant professor of physics at the University of Alabama in Huntsville from 1973 to 1976. At the University of Alabama, he conducted fundamental investigations into the nature of optical absorption in highly transparent solids using infrared spectroscopic and laser calorimetric techniques. During the course of these investigations, he developed many refinements in state-of-the-art techniques to allow for the study of surface and bulk absorption. Before joining the University of Alabama, he was a research physicist at the Naval Research Laboratory from 1972 to 1973, where he conducted similar studies on the optical properties of ionic solids. These included the first high-temperature measurements made on alkali halides using CO_2 laser calorimetry.

At the University of Stuttgart (1970 to 1972) and Northwestern University (1964 to 1970), Dr. Harrington was involved in the study of the optical properties of pure and doped alkaline earth fluorides. These studies spanned the infrared, far infrared, and visible regions using the techniques of infrared spectroscopy, far infrared Michelson interferometry, Raman scattering, and low-temperature thermal conductivity.

JAMES A. HARRINGTON (Continued)

Honors	Special Achievement Award, Naval Research Laboratory (1973); Publication Award, Naval Research Laboratory (1974).
Professional Societies	American Physical Society, Optical Society of America.
Publications	Dr. Harrington has authored over 25 publications in theories of laser calorimetry, lattice dynamics, and optical properties of solids.

ANTONIO C. PASTOR, Member of the Technical Staff, Optical Physics Department, Hughes Research Laboratories.

Education — B.S. (Chemical Engineering), University of Santo Tomas (Manila, Phillipines), 1950; M.S. (Physics), University of Chicago.

Experience — Mr. Pastor joined Hughes in 1966 and has been working on the growth of refractory laser and electro-optic crystals, as well as the design of growth equipments for use in the Hughes reactive atmosphere process for highly transparent alkali halide and alkaline earth fluoride window materials. More recently he has been engaged in similar work on monocrystalline fiber growth of potassium and thallium halides and in the deposition of thin films of inorganic compounds using metallo-organic photoresists. From 1961 to 1968 he was employed at Korad, working particularly on improving the Verneuil method of crystal growth and the development of high temperature crystal preparation methods and the related ceramics and instrumentation techniques. In earlier years, he was an instructor at the University of Santo Tomas and at the Montana School of Mines, and also practiced chemical engineering and operated a commercial laboratory for chemical analysis.

Professional Societies — American Association of Crystal Growers.

Publications — Mr. Pastor is the author of several publications in the Journal of Chemical Physics and the Materials Research Bulletin. He has numerous patents and patent disclosures in the field of crystal growth.

RICARDO C. PASTOR, Senior Scientist, Optical Physics Department, Hughes Research Laboratories.

Education

B.S. (Chemistry), University of Santo Tomas, 1942;
Ph.D. (Chemical Physics), University of Chicago, 1953.

Experience

Dr. Pastor rejoined Hughes in 1968. He has 34 years experience in the study of ionic crystals and electro-optics and laser materials; currently involved in development of surface and bulk materials for solid-state laser hosts, laser windows, thin films, and electro-optical applications. Previously, developed growth process for metal halides and involved in crystal growth and materials evaluation for lasers and nonlinear optics.

For seven years, as Manager of the Chemical Physics Division of Korad Corporation, he directed research in various materials. He was previously Manager of Chemical Physics in the Applied Physics Laboratory of Quantatron, directing programs in new laser materials. When he was employed with Hughes from 1956 to 1961, he performed solid-state chemical research with maser materials. Earlier, built nuclear and electron magnetic resonance equipment in Princeton University to study radiation damage, carried out spectroscopy studies of materials, conducted research in vitamin extraction, and taught chemistry.

Honors

Sigma Xi and Fellow of the Physical Society of London. Recipient of the 1979 L.A. Hyland Award.

Publications

Dr. Pastor is the author of more than 60 papers on paramagnetic resonance, crystal constituent effects, stabilization of dye solutions, and other supporting studies on laser and electro-optic materials in Phys. Rev., J. Chem. Phys., J. Appl. Phys., Rev. Mod. Phys., Mat. Res. Bull., and others. Also author of several patents on a color TV phosphor, a laser head cooling system, preparatioan of low hydroxyl metal halides, purification of alkali earth metal chlorides and bromides, and reactive atmosphere crystal growth methods for metal halides and oxides.

STEPHEN C. RAND, Member of the Technical Staff, Optical Physics Department, Hughes Research Laboratories.

Education

B.Sc. (Physics), McMaster University, 1971;
M.Sc. (Physics), University of Toronto, 1974;
Ph.D. (Physics), University of Toronto, 1978.

Experience

Prior to joining Hughes, Dr. Rand was engaged in light scattering experiments at the University of Toronto, reporting Brillouin measurements in all the rare gas solids and the family of deuterated methane compounds. He spent two years as a World Trade Fellow at IBM Research in San Jose, California. There his research was in the area of optical coherent transients and spectral hole-burning in rare-earth doped crystals. It culminated in the discovery of "magic-angle line narrowing" on an optical transition of Pr ions in LaF_3. Subsequently he spent two years as a research associate in the Stanford University Department of Physics. During this period he worked on radiative pair emission and absorption processes in rare earth materials.

Dr. Rand joined Hughes Research Laboratories in June 1982 and is currently involved in experimental physics research in fiber optics, nonlinear optics, color center lasers, stimulated pair processes and quantum electronics.

Professional Societies

American Physical Society; Optical Society of America.

Publications

Dr. Rand's publications in the above areas include more than 20 papers in Physical Review Letters, Physical Review, Optics Communications, Solid State Communications, Physics Letters and the Canadian Journal of Physics. He has also contributed to several books: Laser Spectroscopy IV (1979), Laser Spectroscopy V (1981), Light Scattering in Solids (1979) and Lasers and Applications (1981).

SECTION 5

INVENTION DISCLOSURES

A. SIMPLE GROWTH METHOD FOR FABRICATION OF SINGLE CRYSTAL HYBRID OPTICAL FIBERS

- Inventor

  Stephen C. Rand

- Summary of the Invention

We describe a technique for embedding optical fiber cores in bulk nonlinear crystals in such a way as to achieve good contact over a significant length and surface area of the fiber. The method uses simple growth in solution of a crystal around a glass fiber to create a hybrid single crystal fiber suitable for nonlinear optics.

B. SATURATED SOLUTION PRESS METHOD FOR FABRICATION OF HYBRID SINGLE CRYSTAL OPTICAL FIBERS

- Inventors

  Stephen C. Rand

  Ricardo C. Pastor

- Summary of Invention

We describe a technique for embedding optical fiber cores in bulk nonlinear crystals in such a way as to achieve good optical contact over a significant length and surface area of the fiber. The method uses pressure to embed a glass fiber in a crystal surface in saturated solution, creating a hybrid single crystal fiber suitable for nonlinear optics.

C. MONOCRYSTALLINE FIBER GROWTH OF CONGRUENTLY MELTING MATERIALS

- Inventors

  A.C. Pastor
  J.A. Harrington
  R.R. Turk
  A.G. Standlee

- Summary of the Invention

This invention consists of a device, and of a method employing that device, for growing single crystal fibers of congruently melting materials from the melt, using as feed stock extruded polycrystalline fibers of the same materials, with or without alteration (reduction or expansion) of the fiber diameter. (This work was initiated under a concurrent ONR contract.)

REFERENCES

1. J. H. Harrington, "Crystalline Infrared Fibers," Infrared Fibers, Proc. of Soc. Photo-Opt. Instr. Eng., 226, 10 (1981).

2. T. L. Brown, General Chemistry, 2nd Edition (C. Merrill Publishing Co., Columbus, Ohio, 1968), p. 340.

3. G. Hewig and K. Jain, J. Appl. Physics, 54, 57 (1983).

APPENDIX A

CRYSTALLINE FIBER OPTICS

(Article prepared for Encyclopedia of Materials Science and Engineering, Pergamon Press, 1982)

J.A. Harrington, Hughes Research Laboratories

Fibers made from crystalline materials have recently been developed for use as infrared (IR) transmissive waveguides and as a media for various nonlinear interactions. As infrared waveguides, single-crystal (SC) fibers may be viewed as a means to improve on the performance of polycrystalline (PC) waveguides extruded from the same crystalline materials. For example, PC KRS-5 (TlBrI) has been extruded into fiber with grain sizes from 3 to 100 μm and losses as low as 0.4 dB/m at 10.6 μm. This PC fiber loss is three-orders of magnitude above the intrinsic loss for KRS-5 at 10.6 μm. By going to SC fibers, the losses should be reduced because scattering and absorptive losses associated with grain boundaries, excessive residual strain induced in the extrusion process, and poor surface quality would be eliminated. In fact, theoretical calculations for a variety of IR crystals indicate that perfect SC fibers should have the ultimate low loss of 10^{-3} dB/km in the 2 to 6 μm region. As a media for nonlinear processes, SC fibers may be used as modulators, signal processors, and as parametric sources. SC fibers of Nd:YAG have been made to lase and could also be used as in-line fiber amplifiers.

A. METHODS OF SC FIBER GROWTH

The methods used to prepare SC fibers fall into two broad categories: edge-defined growth and pedestal growth. In the first category, the fiber material is fed through a capillary which confines and shapes the resultant fiber. The second technique does not use a confining capillary or shaping tube; instead, the feed material is not contained and only the end is melted and drawn into fiber. The basic growth regions are shown in Figure A-1 for either method. In particular, note the key molten (or floating) zone,

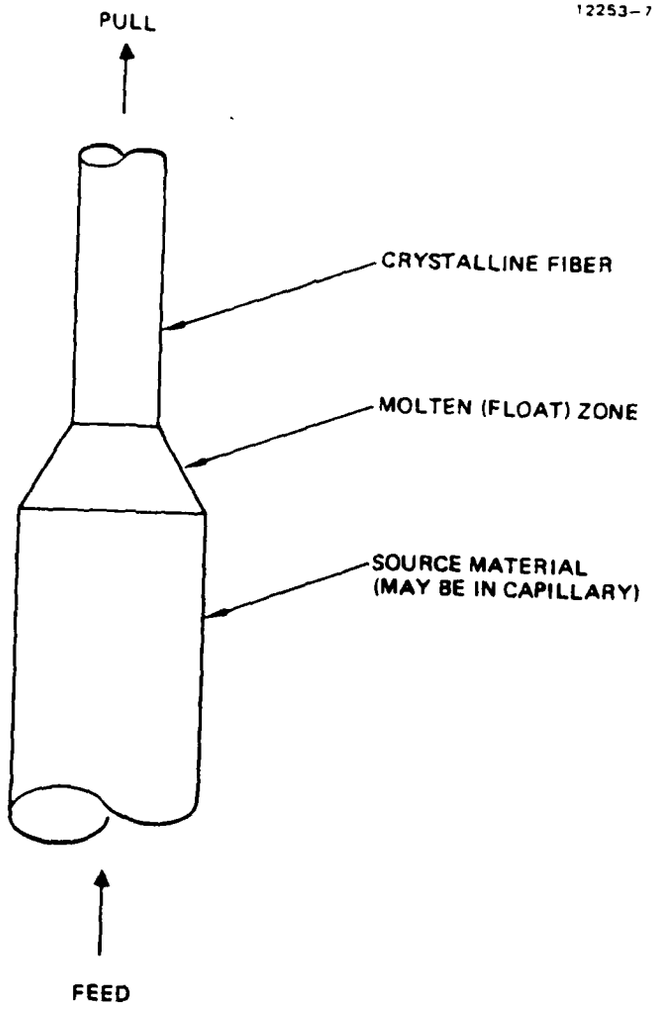


Figure A-1. Growth region for SC fiber fabrication. Source material may be molten material fed from a reservoir through a capillary tube or a solid feed rod. Crystalline fiber can be pulled either up or down.

which generally occurs outside the shaper if one is used. The fiber growth rate and diameter are determined from mass conservation (continuity equation) as

$$d_f^2 V_f = d_s^2 V_s \quad , \tag{A-1}$$

where d_f = fiber diameter, d_s = source rod diameter, V_f = fiber pull rate, and V_s = source rod pull rate. Stable growth is possible for $1 < d_s/d_f < 4$.

Techniques having an edge defining mechanism have been used to grow the lower melting, non-oxide crystals. As seen in Table 1, these crystals are the ductile silver and thallium halides and several alkali halides. These materials are generlly melted in a reservoir which feeds the small capillary or shaper tube made from quartz or platinum. The fiber is pulled vertically from the shaper, either up or down. SC fibers (KRS-5) of up to 2 m in length have been made by this method; long fibers can be fabricated because the source material comes from a molten reservoir.

The pedestal method has been used for higher melting point materials such as the oxides, although the principle should apply to the lower melting salts as well. The top of the source rod is melted, usually with a CO_2 laser, and the fiber is pulled from this small diameter float zone. Very small (30 μm) diameter fibers suitable for highly efficient nonlinear devices have been produced. An advantage of this method is that the surface variations can be less than those prepared with an edge definer because the irregularities in the shaper produce irregular fiber surfaces, whereas in the pedestal method, a smooth surface results if the temperatuare is held constant.

Table A-1. Properties of SC Fibers

SR Fiber Material	Growth Technique	Fiber Diameters μm	Growth Rate cm/min	Loss at 10.6 μm dB/m
CsI	EDG	700-1000	0.5-0.6	12
	EDG	300-800	2	44
KCl	Solution	80-160	months	217
TlCl	EDG	300-800	1	3
KRS-5	EDG	600-1000	0.5-3	--
AgCl	EDG	300-800	3	83
AgBr	EDG	350-750	2 (max)	9
Nd:YAG	Pedestal	50-500	0.1-1	--
Al_2O_3	Pedestal	50-500	0.1-5	--
$LiNbO_3$	Pedestal	50-500	0.1-1	--

B. SC FIBER MATERIALS AND LOSSES

Current losses in SC fibers are high. Table A-1 gives the measured fiber losses at 10.6 μm. Also listed are the range of fiber diameters and the growth rates. The rates are slow (~0.5 cm/min) by comparison to conventional fiber drawing or even PC fiber extrusion. However, they are fast when compared to crystal growth in general. It should be possible, based on Equation (A-1), to increase the growth rate as growth techniques are perfected.

The high losses in the fibers are due to the purity of the starting material and the poor surface quality of the fibers. Pure alkali halides are available (see article on Purification) and will probably be used when the SC techniques are perfected. The surface quality is a major prolem. Surface irregularities result from perturbations in the crystal growth parameters. Specifically, temperature and pulling speed fluctuations lead to fiber diameter variations that cause scattering losses in the fiber. In addition, faceting can occur for certain fiber materials such as the alkali halides. That is, the fibers want to grow with a squarish (alkali halides) or elliptical ($LiNbO_3$) cross-section that is characteristic of Czolchralski-grown boules. As the growth techniques are refined and purer starting materials become available, it is expected that good optical SC fibers will be grown.

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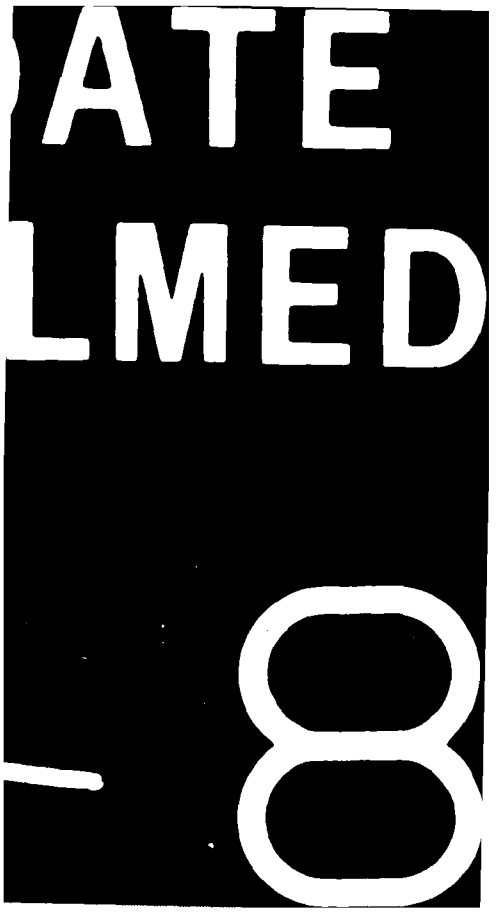
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